AMO Physics with Intense XUV and X-ray Free Electron Lasers

# John T Costello

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# **DCU Laser Plasma-AMO Physics Group**

Laser Plasma/AMO Physics @ NCPST - 6 laboratory areas focussed on pulsed laser matter interactions (spectroscopy/ imaginglparticles)

Principal Investigators (6): John T. Costello, Eugene T. Kennedy (Emeritus), Lampros Nikolopoulos (T), Jean-Paul Mosnier, Paul van Kampen & Paddy Hayden (SFI SIRG PI)

Current Postdocs (3): Dr. Pramod Pandey, Dr. Colm Fallon & Dr. Mossy Kelly

Current PhD students (9 + (1)): Nichola Walsh, Brian Sheehy, Ben Delaney, Stephen Davitt, Hu Lu, Getasew Wubetu, Sri. Inguva, William Hanks, Muhammed Alli & Paul Grimes (Sadaf)

Recent Interns (2012-14): K. Nishant/R. Tejaswi, (LNMIIT, Jaipur), C Hand, (NUIM), S Reddy/R Namboodiri/A Neettiyath (IIT Madras), R Singh/S Gupta (IIT Kanpur), S Howard (Notre Dame), I-M Carrasco Garcia (Malaga)

Recent PhD Grads (2009-2014): Padraig Hough, Conor McLoughlin, Rick O'Haire, Vincent Richardson, Dave Smith, Tommy Walsh, Jack Connolly, Jiang Xi, Leanne Doughty, Eanna MacCarthy, Colm Fallon, Mossy Kelly, D Middleton & Cathal O'Broin

Recent Past Postdocs (2012-2014): Satheesh Krishnamurthy (OU UK), Pat Yeates (Elekta Oncology UK) & Subhash Singh (U. Allahabad).

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### **Collaboration @ FLASH-DESY, Hamburg**

XFEL: P. Radcliffe & M. Meyer

Paris (UPMC): R. Taieb (T) & A. Maquet (T)

PTB (Berlin): M. Richter

DESY (Hamburg): K. Tiedke, S. Düsterer, W. Li, A. Sorokin & P. Juranić, J. Feldhaus

Orsay: D. Cubaynes

Queen's University Belfast: C. L. S. Lewis

Moscow State University : A. N. Grum-Grzhimailo, E. V. Gryzlova, S. I. Strakhova

Crete: P. Lambropoulos (T)

Oulu/GSI: S. Fritzsche (T)

DCU: T. J. Kelly, N. Walsh, E. T. Kennedy, L Nikolopoulos & J. T. Costello

Thanks to AG Photon & AG Machine

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### **Collaboration @ LCLS X-ray FEL (SLAC)**

DESY (CFEL): I. Grguras, M Hoffmann & A. Cavalieri

**DESY (FLASH):** S. Düsterer & J. Feldhaus

DCU: T. J. Kelly, E. Kennedy, V. Richardson, L. Nikolopoulos (T) & J. T. Costello

MPQ/TU-Munich: A. Maier, W. Helml, W. Schweinberger & R. Kienberger

Ohio (OSU): C. Roedig, G. Doumy\* & L. DiMauro

Tohuku University: K. Ueda

Hiroshima University: S. Wada

SLAC: R. Coffee, J. Hastings, C Boestedt, J. Bozek et al.

XFEL Gmbh: P. Radcliffe, T. Tschenscher & M. Meyer

Moscow State University: N. Kabachnik

Thanks to Paul Emma et al.

\*Now at Argonne.

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### Some members of the LCLS collaboration



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# **TIMESCALES - HOW FAST IS FAST ?**



# X-ray – How X-ray is X-ray ?

# Spectral Range: IR to the X-ray



#### Graphic: Courtesy, Prof. David Attwood (Berkeley)

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# What do we want in an X-ray laser ?

The Holy Grail is an X-ray laser with variable pulse duration on the femtosecond to attosecond timescales with tunable wavelength, variable polarisation and high energy per pulse (few 100 µJ to few 10 mJ).....









# How to achieve X-ray lasing.....

- 1. Try to create a population inversion between the well spaced energy levels in highly charged ions -**Optically pumped**, collisional and recombination lasers – expensive and difficult (Livermore, Osaka, Princeton PPL, NRL, Rutherford, etc.).
- 2. Frequency up-conversion by generating high harmonics of an optical laser (low fluxes, hard to scale to X-rays but can yield sub-100 as pulses !!!)
- 3. Make free electron lasers by injecting low emittance electron bunches, supplied by high energy (GeV) linear accelerators into periodic magnetic structures.

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### **SASE-FEL**, Fundamental Principle



1GeV machine  $\gamma$  ~ 2000  $\lambda_{u}$  ~ 2.7 cm /  $\lambda_{laser}$  ~ 6nm

 $\lambda_{\rm L} = \lambda_{\rm u} (1 + {\rm K}^2/2)/2\gamma^2 \qquad \gamma = {\rm E}/{\rm mc}^2$ 

 $K = eB\lambda_u/2\pi mc$ 

Wavelength tunable – by electron beam energy or by tuning the undulator gap Electron bunch slips behind the lightwave by  $\lambda$  per undulator period



### **X-ray Free Electron Lasers (FEL)**





# X-ray Free Electron Lasers (FEL)

#### **LCLS Overview and Specifications**

Injector/Lice 600m er accelerator (SLAC)

Injector/Linac e Beam Transport 227m above ground facility to transport electron beam (SLAC)

Undulator Hall: 170m tunnel housing undulators (ANL)

Electron Beam Dump 40m facility to separate e and x-ray beams (SLAC)

Front End Enclosure 40m facility for photon beam diagnostics (LLNL) Near Experimental Hall: 3 experimental hutches, prep areas, and shops (SLAC/LLNL)

X-Ray Transport & Diagnostic Tunnel; 210m tunnel to transport photon beams (LLNL)



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# X-ray Free Electron Lasers (FEL)

#### **XFEL – Under Construction..... 2016**









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# USPs of XUV & X-ray FELs (XFELs)?

- High flux per pulse typ. 10<sup>13</sup> photons/pulse
- Tunable pulsewidth from 1 to few 100 fs
- Ergo high peak intensity up to few 10<sup>20</sup> W.cm<sup>-2</sup> possible
- Seeded and unseeded modes now possible
- Unseeded bandwidth 0.2 1.0%
- Seeded bandwidth 0.005% (typ.) /  $\lambda/\Delta\lambda \ge 10^4$
- Synchronisation to optical fs lasers relatively easy
- EUV/EUV and X-ray/X-ray pump-probe possible

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# Technology Now.....

So the Holy Grail is now largely realised as the <u>SASE</u> EUV and X-ray FELs at SLAC-Stanford, SCSS & SACLA-RIKEN, FLASH-DESY (+future European XFEL), FERMI@ELETTRA-Trieste, SwissFEL-PSI, Pohang, Shanghai, Dalian, etc.....

Very recently [2012] seeding of LCLS, SCSS and FERMI have resulted in transform limited pulses with  $\Delta\lambda/\lambda$  values of ~ 10<sup>-4</sup>

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# **Ionization in Intense Fields**

#### 1. Rudiments of ionization processes in intense laser fields

Sfinsp

- 2. Photoionization experimental setups (FLASH & DESY)
- 3. One colour two photon ionization

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- 4. Two colour Ionization physics and characterisation
- 5. Some future perpectives

# **The Atomic Photoelectric Effect**



# What happens as the laser intensity (field strength) grows ?



# How can you determine in which regime the interaction resides ?

$$\gamma = \sqrt{\frac{IP}{2U_p}}$$
 Keldysh Parameter  
IP = Ionization Potential  
Up = Ponderomotive Pot.

$$U_P = 9.3 \times 10^{-14} I (W cm^{-2}) \lambda^2 (\mu m) eV$$

\*L V Keldysh, Sov.Phys-JETP 20 1307 (1965)

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**Potential** 



# **Keldysh - Ionization Regime**

Multiphoton IonizationTunnel IonizationField Ionization $\gamma >> 1$  $\gamma \sim 2$  $\gamma << 1$ 

Example: <u>Helium</u> in intense laser fields

For Ti-sapphire laser: 800 nm,  $10^{15}$  Wcm<sup>-2</sup>,  $\gamma \sim 0.45$  (TI/FI regime) For an EUV laser: 8 nm,  $10^{15}$  Wcm<sup>-2</sup>,  $\gamma \sim 45$  (MPI regime)

So for EUV lasers, multi-photon ionization is the primary processs and will involve *few photons* and *potentially few electrons* 





# **USPs of XUV & XFELs in AMO Physics ?**

- *Ultra-dilute* targets
- Photo-processes with *ultralow cross-sections*
- **Pump and probe** experiments (EUV + EUV or EUV + Opt.)
- Single shot measurements
- *Few-photon* single and multiple *ionization processes*
- NB1: Makes *inner-shell* electrons key actors in non-linear processes for the first time
- NB2: Re-asserts *primacy of the photon* over field effects !



# **Experimental Setups (DESY & SLAC)**

- 1. Rudiments of ionization processes in intense laser fields
- 2. Photoionization experimental setups (FLASH & LCLS)
- 3. One colour two photon ionization
- 4. Two colour Ionization
- 5. Some conclusions



# **Photoelectron Spectroscopy @ FLASH**

#### **Experimental Layout at FLASH - (EU-RTD)**



# **AMO PES Chamber at LCLS**

#### **Rendered Image:**

High Field Chamber (AR-ETOF) and Diagnostics (MBES) Chamber



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# Two Photon Ionization (TPI) of Xe and Kr atoms in an Intense Field

- 1. Rudiments of ionization processes in intense laser fields
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### Non-linear processes in the EUV & X-ray

Question. What is the simplest experiment you can carry out in non-linear optics ? Answer. Either two-photon absorption (TPA) or second harmonic generation (SHG)......



### **Motivation - Xe TPI in intense EUV fields**

Sorokin, Richter et al., PTB, PRL 2007 – Ion Spectroscopy !!



# Xe + hv (93 eV) - Xe<sup>+</sup>(4d<sup>-1</sup>) + e<sup>-</sup> (~25 eV)



# Xe + 2hv (93 eV) - Xe<sup>+</sup>(4d<sup>-1</sup>) + e<sup>-</sup> (~ 118 eV)

Now ramp up the intensity to > 10<sup>15</sup> W.cm<sup>-2</sup>.....

•Using MBES, first evidence of two photon *inner* shell ionisation, (in this case) of 4d electron – Xe +  $2hv \rightarrow Xe^+ 4d^9 + e^-$ 

'Retardation field' applied to suppress low KE electrons (one photon processes)
– hence electrons detected are due solely to multiphoton events

•Energetically – 2 × (93) eV – 118 eV = 68 eV

•Yield scales quadratically,  $n=1.95 \pm .2$ 





Insp

DC

### **Resonant 2-photon, 3d Excitation of Kr**

1. To date we have looked at a **non-resonant** two photon process (sort of ATI really)

2. FELs are wavelength tunable - one can also explore resonant two photon processes
Kr 3d<sup>10</sup>4s<sup>2</sup>4p<sup>6</sup> (<sup>1</sup>S<sub>0</sub>) + 2 x hv (46 eV) -> 3d<sup>9</sup>4s<sup>2</sup>4p<sup>6</sup>4d (J=0,2)

i.e., 3d - 4d two photon resonant excitation



# **Kr - Resonant Two Photon Excitation**

- 1. Kr 3d<sup>10</sup>4s<sup>2</sup>4p<sup>6</sup> (<sup>1</sup>S<sub>0</sub>) + 2 x hv (46 eV) -> 3d<sup>9</sup>4s<sup>2</sup>4p<sup>6</sup>4d (J=0,2) i.e., 3d - 4d two photon excitation
- Of course there is a direct ionization path and the usual interference results - manifested as asymmetric resonance profiles (Fano/ Fano-Mies)
- But here the 3d<sup>9</sup>4s<sup>2</sup>4p<sup>6</sup>4d (J=0,2) resonance undergoes Auger decay to Kr<sup>+</sup> on a femtosecond timescale - similar to the FLASH pulse duration - so competition between excitation and decay (ergo, in addition to simple ATI, this case makes for an intriguing, problem for theory)..

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#### *hv* = 46 eV (~27 *nm*)



Meyer et al., PRL 104 213001 (2010)

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# Kr (3d<sup>9</sup>4d) 2 Photon Resonance Auger

MBES Photoelectron spectrum - ~  $5 \times 10^{14} \text{ W.cm}^{-2}$ 



EXTATIC Phys Rev Lett **104** Art. No. 213001 (2010) **NCP** 





# **Summary - One Colour**

- Xenon Demonstration of an 'above threshold absorption-ionization' twophoton process involving an *inner shell electron*.
- It is clear that the although single photon ionization processes dominate, they are sufficiently important at high irradiance that, for a given intensity, much higher ionization stages can be reached compared to optical lasers.
- The strength and the nature of the 4d → εf resonance may open up, at high irradiance, additional ionization channels, namely the simultaneous multiphoton / multi-electron from the inner 4d shell, 'inside-out ionization' or 'peeling the onion from the inside out'
- Kr was first step on the road to resonant NL processes with EUV/Xrays.... **REMPI at X-rays.**

*Xe - Richardson et al. PRL (July 2 – 2010), Kr - Meyer et al., PRL (May 28 - 2010)* 







# XUV (X-ray) + IR Ionization

- 1. Rudiments of ionization processes in intense laser fields
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# Atoms in Intense Superposed X-ray + IR Laser Fields

## Main objective

Study the effect of X-ray pulse width on fundamental photoionization processes in intense and ultrashort ionizing (X-ray) and dressing (Optical / IR) laser fields

#### **Two Extremes:**

X-ray pulse duration is 'many' optical cycles X-ray pulse duration is less than ½ optical cycle







# Atoms in 'Long' XUV (X-ray) + IR Fields

Sideband number/intensity depend strongly on XUV/NIR overlap  $\Rightarrow$  by comparison with theory we are able to determine relative time delay to better than 100 fs



Ultrafast XUV-modulated optical-reflectivity methods
 Gahl et al., Nature Photonics 2 165-169 (2008)
 Maltezopoulos et al., New J Phys 10 Art. No. 033026 (2008)

NIMA **83**, 516-525 (2007) Appl. Phys. Lett **90** 131108 (2007) 2. TEO
A. Azima et al., APL,
94 144102 (2009)

inspire



# Streaking.....

# Two colour photoionization experiments – The Atomic X-ray Streak Camera

The key diagnostic in ultrafast laser and optical physics is the Streak Camera. It is essentially an optical oscilloscope where the input channel is a photocathode as opposed to the usual direct electrical BNC input.....



Hamamatsu Synchroscan at DCU

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# Streaking.....

#### Streak Camera Operation – Courtesy Hamamatsu Corp.



# Atoms in 'Short' XUV (X-ray) + IR Fields



# Atoms in 'Short' XUV (X-ray) + IR Fields

Experimental realisation - optical delay line used to sweep an attosecond X-ray pulse, focused into a gas jet, past an intense fs optical laser field measuring the photoelectron kinetic energy at each point => *the Electric Field of the optical laser revealed......* 



M Drescher et al. Nature 419, 803–807 (2002) / R Kienberger et al., J. Mod. Opt 52 261-275 (2005)

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### Measurement of few fs pulses @ LCLS

#### **Experimental Layout at LCLS**



### **Measurement of few fs pulses @ LCLS**

LCLS low current/slotted spoiler/ few fs mode -Data still under analysis.....

Process. Ne + hv (1.8 keV) -> Ne<sup>+</sup> (1s<sup>-1</sup>) + e<sup>-</sup> +  $I_L$  (10<sup>14</sup> W.cm<sup>-2</sup>)

Essentially mapping time (fs) to energy in (eV) allows one to measure X-ray (and EUV) pulse widths to attosecond accuracy provided the X-ray (EUV) pulse width is less than one one half cycle of the optical laser in duration !!



### Sub-femtosecond pulses @ LCLS

#### 800 as X-ray pulse !!

Process. Ne + hv (1.8 keV) -> Ne<sup>+</sup> (1s<sup>-1</sup>) + e<sup>-</sup> +  $I_{L}$  (10<sup>14</sup> W.cm<sup>-2</sup>)



### Single Cycle THz Streaking @ FLASH

#### Femtosecond Atomic Streak Camera

Generate single (picosecond) cycle pulse using optical rectification of Ti-Sappire laser pulses – field ~ 50MV/m maximum



#### Schematic layout of the THz Streaking Experiment at FLASH

EXTATIC Nature Photonics 6 pp852-857 (2012)





# Single Cycle THz Streaking @ FLASH 47

#### Femtosecond Atomic Streak Camera

Generate single (picosecond) cycle pulse using optical rectification of Ti-Sappire laser pulses – field ~ 50MV/m maximum



#### **Principle of the experiment**

Attosecond Photoelectron Streaking showing how the Efield of a few cycle fs laser pulse can be mapped – MPI-Q.

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### Single Cycle THz Streaking @ FLASH

#### A Cavalieri et al. from CFEL, DCU, XFEL & DESY



Single cycle THz Photoelectron Streaking showing how the Efield of a single cycle ps laser pulse can be mapped





Jitter measurements on 50 consecutive streak traces



Nature Photonics 6 pp852-857 (2012)



### **LCLS - Single Cycle THz Streaking**

#### A Cavalieri et al. from CFEL, DCU, XFEL & SLAC



If the dispersed bunch is intercepted by a 'V-shaped' vertical slot, then **the emittance of the all but TWO small parts in space (time) of the bunch is 'spoiled'** -=> 2 X 'few fs' pulses of variable separation result.

P Emma et al., PRL 109 254802 (2012)



### **NEW !! All Optical Synchronisation - FLASH**

#### A Cavalieri et al. from CFEL, DCU, MPI (SDM), SLAC & XFEL



# **Measuring Polarisation of XFELs**

T Mazza et al. (XFEL GmbH, DESY, FERMI@ELECTTRA, DCU, MSU, etc) Theory - Kazansky, A. K., Grigorieva, A. V. and Kabachnik, N. M. Circular Dichroism in Laser-Assisted Short Pulse Photoionization. Phys. Rev. Lett. 107, 253002 (2011).





- 1. Rudiments of ionization processes in intense laser fields
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# **Imaging single molecules !!!**







Single shot dynamic coherent diffraction imaging on femtosecond timescales - soon to be used in single biomolecular imaging to make molecular movies !!!

#### Cf: CFEL, DESY, LCLS and PULSE-Stanford

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# **X-Ray Lasers - Future**

### Speculation

Ordinary X-rays are used in Diagnostics (Images) and Therapeutics (Cancer/Radiography).

X-ray lasers add the possibility to make 3D images (holograms) of the molecules that cause diseases and follow them on a femtosecond timescale as they do so !!

Molecular (Nanomedicine).....

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# Self - Seeded FELs, e.g., LCLS.....



Single-Shot Spectra



**Multi-Shot Averaged Spectra** 



Lutman et al., PRL 113 Art. No. 254801 (2014)/Amann et al. Nature Photonics 6, 693 (2012)

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# In Conclusion

- 1. To date we have looked only at one and two colour nonresonant photoionization processes
- 2. Now FELs seeded and easily tunable we can explore resonant processes where inner shell electrons dominate

Next steps (XFEL Technology): X-CPA

XFELs are finally becoming real lasers – truly monochromatic, fully phase coherent, collimated..... If it can be done with an optical laser – now propose it for XFELs....



### **Regular Articles**

- 1. Spectroscopic characterization of vacuum ultraviolet free electron laser pulses, Optics Letters 31 1750 (2006)
- 2. *Two-color photoionization in xuv free-electron and visible laser fields*, Phys. Rev. A **74**, Rapid Communications, Art. no. 011401 (2006)

3. *Single-shot characterization of independent femtosecond extreme ultraviolet free electron and infrared laser pulses*, Appl. Phys. Lett **90**, Art. no. 131108 (2007)

4. Operation of the Free Electron Laser FLASH in the water window, Nature Photonics 1 336 (2007)

5. An experiment for two-color photoionization using high intensity extreme-UV free electron and near-IR laser pulses, Nucl. Inst. Methods in Res. A **583** pp516-525 (2007)

- 6. Polarization control in atomic 2-color above threshold ionization, Phys. Rev. Letts 101 Art. no. 193002 (2008)
- 7. Time-resolved pump-probe experiments beyond the jitter limitations at FLASH, Appl. Phys. Letts 94 Art. no. 144102 (2009)
- 8. Two-Photon Excitation and Relaxation of the 3d-4d Resonance in Atomic Kr, Phys. Rev. Letts 104 Art. no. 213001 (2010)
- 9. Two-photon inner-shell ionization in the extreme-ultraviolet (XUV), Phys. Rev. Letts 105 Art. no. 013001 (2010)
- 10. Two-color experiments in the gas phase at FLASH, J. Electron. Spec. Relat. Phenom. 181 pp111-115 (2010)
- 11. *Femtosecond x-ray pulse length characterization at the LCLS FEL*, New J. Phys. **13** Art. no. 093024 (2011)
- 12. Theory of ac-Stark splitting in core-resonant Auger decay in strong x-ray fields, Phys. Rev. A 84 Art. no. 063419 (2011)
- 13. Angle-resolved electron spectroscopy of laser-assisted Auger decay induced by a few-fs x-ray pulse, Phys. Rev. Letts. 108 Art. no. 063007 (2012)

14. Atomic photoionization in combined intense XUV free-electron and infrared laser fields, New J. Phys. **14** 043008 (2012)

- 15. *Dichroism in the above-threshold two-colour photoionization of singly charged neon*, J. Phys. B: At. Mol. Opt. Phys. **45** 085601 (2012)
- 16. Controlling core hole relaxation dynamics via intense optical fields, J. Phys. B: At. Mol. Opt. Phys. 45 141001 (2012)
- 17. Ultrafast X-ray pulse temporal characterization for free-electron lasers, Nature Photonics 6 852-857 (2012)
- 18. *Determining the polarization state of an XUV free-electron laser beam using atomic circular dichroism*, Nature Communications **5**, 3628 (2014)
- 19. *Measuring the temporal structure of few-fs FEL X-ray pulses directly in time domain*, Nature Photonics **8**, 950 957 (2014)
- 20. Femtosecond, all-optical synchronization of an X-ray free-electron laser, Nature Communications 6, Article no. 5938 (2015)











### **Review Articles**

1. Photoionization experiments with the ultrafast XUV laser FLASH J. T. Costello, J. Phys. Conf. Series 88 Art No. 012057 (2007)

#### 2. Experiments at FLASH

C. Bostedt, H. N. Chapman, J. T. Costello, J. R. Crespo Lopez-Urrutia, S. Duesterer, S. W. Epp, J. Feldhaus, A. Foehlisch, M. Meyer, T. Mšller, R. Moshammer, M. Richter, K. Sokolowski-Tinten, A. Sorokin, K. Tiedtke, J. Ullrich and W. Wurth, *Nucl. Inst. Meth. in Res. A* 601 108-122 (2009)

3. Non-linear processes in the interaction of atoms and molecules with intense EUV and X-ray fields from SASE free electron lasers (FELs)
N. Berrah, J. Bozek, J. T. Costello, S. Duesterer, L. Fang, J. Feldhaus, H. Fukuzawa, M. Hoener, Y. H. Jiang, P. Johnsson, E. T. Kennedy, M. Meyer, R. Moshammer, P. Radcliffe, M. Richter, A. Rouzee, A. Rudenko, A. Sorokin, K. Tiedtke, K. Ueda, J. Ullrich and M. J. J. Vrakking, *Journal of Modern Optics* 57 1015-1040 (2010)

4. Two-colour experiments in the gas phase
M. Meyer , J. T. Costello , S. Düsterer , W. B. Li and P. Radcliffe
J. Phys. B: At. Mol. Opt. Phys. 43 Art No. 194006 (2010)

5. Two-Color Experiments in the Gas Phase at FLASH M Meyer et al., J Electron. Spec. Relat. Phenom **181**, 111-115 (2010)

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# **'EXTATIC' – Extreme-ultraviolet &** X-ray Technology And Training for Interdisciplinary Cooperation

# http://www.extatic.eu



# Funding

Higher Education Authority – Programme for Research in Third Level Institutes (IV and V)

Science Foundation Ireland – Investigator Programme – 12/IA/1742 & 07/IN.1/I1771

Irish Research Council (PhD Scholarships / Postdoctoral Fellowships)

EU FP7 Erasmus Mundus Joint Doctorate 'EXTATIC' - FPA 0033-2012 and Marie Sklowdowska Curie – Proj. No. 628789

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